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F. Courtine, S. Sanzelle, Thierry Pilleyre, Didier Miallier. Calibration of a germanium well-detector using  $^{60}\text{Co}$ : the effects of the angular correlation of the two gamma rays emitted in cascade, quantified with Monte Carlo simulations. *Radiation Measurements*, 2014, 61, pp.78-82. 10.1016/j.radmeas.2013.11.007 . in2p3-00176273

**HAL Id: in2p3-00176273**

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Submitted on 3 Oct 2007

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Calibration of a germanium well-detector using  $^{60}\text{Co}$ : effects of the correlation between the two gamma rays emitted in cascade, quantified by means of a Monte Carlo simulation.

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## 1 - Introduction

Radiation sources of  $^{60}\text{Co}$  are commonly used for the calibration of germanium gamma spectrometers, both for energy and for efficiency (e.g., Allisy et al., 1994). Decay of this nuclide gives rise to two gamma rays at 1173.2 keV ( $\gamma_1$ ; intensity :  $\rho_1 = 0.9985$ ) and 1332.5 keV ( $\gamma_2$ ;  $\rho_2 = 0.9998$ ) respectively, which follow each other within around  $10^{-12}\text{s}$  (data from the National Nuclear Data Center, Brookhaven National Laboratory). Because such duration is significantly shorter than the one that is required by the electronic setup to gather a pulse, i.e., in the order of some tenths of microseconds, the energy deposition of both gamma rays, if any, can be considered as simultaneous in the detector. Then, discrimination between both photons is not possible as only the sum of the deposited energies will be recorded. Such effect, known as “cascade effect” or “coincidence-summing” will result in a deficit of counts in the two corresponding photopeaks. The apparent counting efficiency measured for one of the peaks is lower than the efficiency that would be evaluated at the same energy, in the same conditions, with a single gamma ray. For overcoming this drawback in evaluating the counting efficiency,  $\varepsilon_E$ , vs photon energy, it is possible to use the sum peak, i.e., the observed peak at 2505 keV, which is due to the summation of the two gamma rays when totally absorbed, in coincidence, by the detector.

On the other hand,  $\gamma_1$  and  $\gamma_2$  are correlated to each other as concerns direction. That is  $\gamma_2$  is not emitted randomly, the probability that it is emitted in the same direction - parallel or anti-parallel - as  $\gamma_1$  being 17% higher than emitted at  $90^\circ$ . The phenomenon has been theoretically studied by Biedenharn and Rose (1953), and confirmation was brought by several experiments, e.g., by Klema and McGowan (1953).

In the perspective of using  $^{60}\text{Co}$  for calibrating a well germanium gamma spectrometer, we wondered whether the correlation could induce in the detector a different collection of photons than in the case of random emissions. Such question has already been addressed by Roteta et al. (1996), which focused their investigation on the single peaks for various nuclides, with three different measurement geometries: planar Ge, co-axial Ge and well NaI.

The expressions giving the corrections induced by the correlation were calculated on the basis of the efficiency of the detectors and on some nuclear parameters attached to the nuclides of interest. A Monte Carlo approach was used for averaging the angular correlation distribution. As a result, they found that the effect of angular correlation on coincidence summing corrections can be neglected if the source is measured either in contact with the detector or at large distances. But the correction remains very weak in any case, i.e., lower than 1%. Complementary exploration of the question was further performed by García-Torano et al. (2005), which evaluated the corrections for coincidence only (i.e., without the angular correlation) on the single peaks for a volume source. They used the simulation package PENELOPE for this purpose.

In the present work, we focused our interest on the sum peak, because it can be used for calibration, in the case the coincidence corrections cannot be accurately evaluated.

Actually, the apparent counting efficiency for the sum peak is equal to the squared efficiency for a single gamma ray at 1250.3 keV (see Appendix). Accordingly,  $\varepsilon_{1,25}$  is derived from equation [1]:

$$\varepsilon_{1,25} = \sqrt{\frac{N_{sum}}{A\rho_1\rho_2.\Delta t}} \quad (1)$$

$N_{sum}$  is the number of counts in the sum peak.  $\Delta t$  (s) is the effective counting duration, i.e., the real time minus the dead time, which actually was negligible with a low activity source.  $A$  (Bq) is the activity of the source.

## 2 - Simulation of the detector response to gamma rays

### 2 - 1 - Testing the validity of the simulation

We considered the spectrometer used in our group. It is based on a 193 cm<sup>3</sup> intrinsic germanium crystal, of the well geometry, encapsulated in a Kryal (aluminium alloy) shield (Canberra, Model GCW3523, Fig.1). Such detector is adapted to measurements of low energy gamma rays emitted from a small source inserted inside the well, which has thin walls on purpose. Alternatively, it is also available for larger external sources, e.g., liquids and powdered samples contained in flat containers (co-axial geometry) or Marinelli beakers. Therefore, it should be calibrated for efficiency both as regards internal (i.e., in the well) or external sources. In this view, we have been using the Monte Carlo code Geant 4, which was initially designed for the needs of high energy physics research (Agostinelli et al., 2003). The

*Low Energy Package* of Geant4 was used. It allows tracking the interaction between photons, electrons and matter down to 250 eV in energy. The code can simulate all the processes up to several hundreds of GeV, thus including those that are significant in the energy range of interest, i.e., [10 keV - 3000 keV]. That is principally Compton and photoelectric effects for photons, and, to a lesser extent, pair production. It can record all the energy losses, at each interaction, so that they can finally be processed for analysis. Basically, in the simulation, the trajectory of the particles is divided in multiple steps. At every stage, the direction, the kind of interaction and the energy that is deposited are selected according to the corresponding probability laws. When becoming weaker than 250 eV after several interactions, the remaining energy is considered to be entirely and immediately deposited in the material. Collection and analysis of the data was made by means of Root, which is a software developed at the CERN (Genève, Sw.) in the frame of high energy physics by Brun and Rademakers (<http://root.cern.ch>).

(Fig.1 here)

The principle of the approach was (i) generate mono-energetic photons from a localized point source, (ii) simulate their interactions in the whole set, including the crystal with its mounting and shielding, and then (iii) cumulate the total energy deposited in the crystal for each single photon.

The efficiency was defined as the ratio of the number of photons,  $N$ , which have deposited all their energy in the crystal, by the number,  $N_0$ , of the generated photons:

$$\varepsilon = \frac{N}{N_0}$$

Such efficiency depends on the energy and on the localisation of the source for a given detector.

In the simulation, the minimal number of generated photons was set so that the minimal number of counts in the sum peak was  $10^4$ . It implied  $10^7$  photons generated inside the well, and  $10^9$  outside. The statistic uncertainty was estimated by processing ten independent runs for each position of the source. This resulted in uncertainties in the range [0.2 %- 2 %]. This is highly time-consuming for an ordinary PC computer (respectively 7.5 h and 31 days). However, the duration was divided by  $\sim 10$  by using several parallelized computers (Centre de Calcul IN2P3, Villeurbanne, France).

For validating the results of the simulation, it was necessary to compare them to experimental data obtained with a single gamma ray. For that purpose, we used the 661.7 keV line from a

$^{137}\text{Cs}$  reference point source, the activity of which was known to  $\pm 2\%$  ( $2\sigma$ ). The experimental efficiency was evaluated as:

$$\varepsilon_{0.66} = \frac{N_m}{0.85 A \Delta t}$$

0.85 is the intensity of the 661.7 keV line of  $^{137}\text{Cs}$ .  $N_m$  is the total number of counts that are measured in the peak; it is evaluated using the *Genie 2000* Canberra software.

In this work, we have considered a point source located on the axis of the crystal, either inside or outside the well. The efficiency was evaluated at various positions of the source on this axis, both using the experiments and the simulation. In the simulation, the dead layers of germanium were taken into account with the thicknesses that were given by the manufacturer, i.e., 1mm for the external wall, 400 $\mu\text{m}$  for the internal one and 100 $\mu\text{m}$  for the horizontal surface (not reported on Fig.1). Then, the calculated and experimental efficiencies were plotted against the distance from the bottom of the well (Fig.2).

(Fig.2 here)

As a result, the two curves exhibited the same shape and the efficiencies differed by 7% at the maximum from each other. It was then concluded that the model was globally valid, although it had to be adjusted. Such adjustment was subsequently performed on the basis of a feedback from experiments (Courtine, unpublished these, 2007). However, since it would necessitate further developments for justification, the adjusted model was not used in the work presented here. Actually, not accounting for the refinements was of negligible consequences on the effects discussed in the present paper because we deal with relative variations which are controlled by the basic geometry of the crystal.

## 2 - 2 - Simulating the cascade and the correlation

In order to simulate the cascade, pairs of photons of 1.17 and 1.33 MeV were created. For each pair, the sum of the energies deposited into the crystal by the two photons was recorded. It allowed plotting the histogram of counts vs energy (by pair). The efficiencies were derived from the number of counts in the peaks. For sake of comparison, calculations were processed either with the correlation or without it.

The correlation can be expressed as:

$$W(\theta) = 1 + a \cos^2 \theta + b \cos^4 \theta$$

$\theta$  is the angle between the photons directions. The coefficients  $a$  and  $b$  have been theoretically evaluated by Biederharn and Rose (1953), who got:  $a = 0.125$ ,  $b = 0.0417$ . Satisfactorily comparable results were experimentally obtained by Klema and McGowan (1953):  $a = 0.131 \pm 0.017$ ,  $b = 0.024 \pm 0.017$ . It can be verified that,  $W(0^\circ) = W(180^\circ) = 1,17$ , whereas  $W(90^\circ) = W(270^\circ) = 1$ . That is, the probability that the direction of the second photon is parallel or anti-parallel to the first one is 17% higher than for orthogonal.

Although Geant4 can simulate the disintegration of a given nuclide like  $^{60}\text{Co}$  by means of the Radioactive Decay Module (Truscot, 2000), the code does not include the angular correlation. Therefore, we had to implement this function in our program. In Geant4, the momentum of the gamma rays  $\vec{P}_i$  is drawn at random. For a couple of emissions, this induces the generation of  $\vec{P}_1$  and  $\vec{P}_2$ . Those data can be extracted from the simulation. Then, accounting for the relation [5] which is valid if  $P_1$  and  $P_2$  are normalised,  $\cos(\theta)$  can be calculated for every couple of draws.

$$\vec{P}_1 \cdot \vec{P}_2 = \cos(\theta)$$

From knowledge of  $\cos(\theta)$  it is possible to impose that  $\theta$  is distributed according to the relation [4] before allowing the program to process the data. It was done here by using the technique “accept-reject” developed by Von Neuman (Nougier, 1985, Press et al., 2005). By such means we made sure that the generated pairs of photons followed the given distribution. In order to verify that the method was correct, it was preliminary tested successfully in a simple geometry. The correlation was then applied to the germanium detector geometry. Simulations were made for a source located on the main axis, either in the well or outside.

(Fig. 3 here)

### 3 - Results and discussion

Fig.3a represents a gamma spectrum generated by the method presented above. It can be verified that it shows the same features as an experimental spectrum. (Fig.3b).

In a first step, we calculated the counting efficiency for the two peaks with - and without the cascade effect. Then, for evidencing the possible effect of the correlation, we compared the counting rates in the three peaks with and without correlation (Tab.1).

(Table 1, here)

The results can be considered from two different points of view: in the well ( $z \leq 55$  mm) or outside.

For a source situated in the well, the cascade will affect significantly the counting rate for the two peaks. For example, the lack of counts at 1.17 MeV ( $z = 40$ mm), compared to a single gamma ray at the same energy is as high as 29 %. As a consequence, the experimental efficiency which will be measured with  $^{60}\text{Co}$  cannot be used for assessing the detector efficiency at 1.17 MeV. On the other hand, the counting rate for the sum peak is not affected by the correlation, the ratio (with and without correlation) being not significantly different from 1 (Tab. 1). Then, there is no significant bias in using the apparent efficiency of the sum peak for evaluating the detector efficiency at 1.25 MeV with equation (1).

Problems arise when the source is placed above the crystal container, because the solid angle between the source and the crystal becomes significantly lower than  $4\pi$ . Up to  $z = 100$  mm at least, the cascade effect still has to be accounted for when using the 1.17 or 1.33 MeV peaks. On the other hand, it appears that the actual sum peak (i.e., with the correlation) is higher by more than 3% than the sum peak which would exist in the hypothesis of no correlation (Tab. 1 and Fig. 4). In such situation the sum peak is of no use either, in evaluating the detector efficiency for a given energy (1.25 MeV), unless the relevant correction has been calculated. It should be outlined that the region where the difficulty is the most important, i.e. just above the upper surface of the detector, corresponds to the one of common use for measuring large sources. It concerns, e.g., unknown samples of contaminated material enclosed in a flat box.

In any case, the correlation effect on the single peaks appears to be lower than 1%, in agreement with the results obtained by Roteta et al. (1996) with another approach.

#### 4 - Conclusions

Roteta et al. (1996) have demonstrated that the angular correlation between two gamma rays emitted in cascade by a nuclide generally does not induce a significant bias in the counting efficiency for the single peaks. However, the problem remains of evaluating the lack of counting in such peaks, due to the cascade effect itself. Assessing this deficit accurately is not simple, because it requires a good knowledge of the detector geometry and electronics and it implies the availability of a simulation code along with a powerful computer (see, e.g.,

García-Torano et al., 2005). Then, the simplest way to overcome the corresponding difficulties in evaluating the counting efficiency for a given source will consist in placing the source far away from the detector so that the probability that the two photons of a pair interact in the detector crystal becomes negligible. Unfortunately, this results in a low efficiency.

Alternatively, for some nuclides like, e.g.,  $^{60}\text{Co}$ , a sum peak can be used because its apparent efficiency is simply related to the efficiency for a single peak at a given energy (Eq. 1). It has been demonstrated in the present paper, by means of the GEANT4 simulation code, that the sum peak is not biased by the angular correlation only for a source inserted in a well inside the detector crystal. But, when the source is placed outside, in the vicinity of the detector, the sum peak is significantly affected by the angular correlation and this effect is enhanced with increasing distance.



	<b>h (mm)</b>	<b>1</b>	<b>20</b>	<b>40</b>	<b>60</b>	<b>70</b>	<b>80</b>	<b>90</b>	<b>100</b>
<b>Cascade without correlation</b>	<b>R<sub>0.1.17</sub></b>	0.58	0.59	0.71	0.88	0.92	0.94	0.96	0.97
	<b>R<sub>0.1.33</sub></b>	0.56	0.57	0.70	0.87	0.91	0.94	0.95	0.97
<b>Cascade with correlation</b>	<b>R<sub>1.17</sub></b>	1.001 ± 0.002	0.999 ± 0.002	1.000 ± 0.002	0.997 ± 0.002	0.996 ± 0.003	0.997 ± 0.003	0.997 ± 0.003	0.997 ± 0.004
	<b>R<sub>1.33</sub></b>	0.999 ± 0.002	0.999 ± 0.002	0.999 ± 0.002	0.997 ± 0.002	0.996 ± 0.003	0.996 ± 0.003	0.997 ± 0.003	0.997 ± 0.004
	<b>R<sub>sum</sub></b>	1.003 ± 0.003	0.998 ± 0.005	1.00 ± 0.01	1.029 ± 0.014	1.048 ± 0.002	1.061 ± 0.003	1.078 ± 0.004	1.079 ± 0.004

Table 1. R<sub>0</sub>: ratio of the number of counts in the peak for a photon affected by the cascade effect, without taking into account the correlation, by the number of counts in the peak for a single photon (statistical uncertainty not quoted for sake of clarity, is lower than 0.3 %). R: ratio of the number of counts in the peak with the correlation by the number of counts in the peak without the correlation. Calculations by means of Geant4 were performed at 1.17MeV, 1.33 MeV and for the sum peak. h is the distance between the <sup>60</sup>Co point source and the bottom of the well.

Figure captions

Figure 1

Axial cross section (simplified) of the detector. Dimensions in mm.

G : germanium; H: copper holder; K: Kryal shielding.

Figure 2. Counting efficiency vs distance from the bottom of the kryal well for the 662 keV gamma rays emitted by a point source of <sup>137</sup>Cs: experimental results and results calculated using GEANT4.

Figure 3. Gamma ray spectra for  $^{60}\text{Co}$ , for a point source situated on the detector axis, at 40mm from the bottom of the Kryal well. A: measured using a reference source; B: simulated using the Monte Carlo code Geant4.

Figure 4. Ratio of the number of counts in the sum peak, with the correlation by the number of counts in the sum peak without the correlation (data from Tab. 1). Calculations performed with a point source at a distance h from the bottom of the Kryal well.

APPENDIX: Evaluation of the counting efficiency for the  $^{60}\text{Co}$  sum peak.

The number of counts in the sum peak,  $N_{sum}$ , is related to the counting efficiencies  $\varepsilon_1$  and  $\varepsilon_2$  at respectively 1.17 Mev and 1.33 Mev, by the relation:

$$N_{sum} = \varepsilon_1 \varepsilon_2 A \rho_1 \rho_2 \Delta t$$

In the given range of energy, it can be experimentally verified that the efficiency is linked to the energy by a linear relation, in a log scale, as:

$$\text{Ln}(\varepsilon) = K \text{Ln}(E) + \text{Ln}(B)$$

K ( $K < 0$ ) and B are factors depending on the experimental conditions.

Such relation can be expressed as:

$$\varepsilon = BE^k$$

Then:

$$\varepsilon_1 \varepsilon_2 = B^2 (E_1 E_2)^k = B^2 (1.2503)^{2k}$$

with:  $E_1 E_2 = 1.563289 = (1.2503)^2$  (E in MeV)

Actually,  $B^2 (1.2503)^{2k}$  is the squared efficiency at 1.2503 Mev, that is  $(\varepsilon_{1.25})^2$ , then:

$$\varepsilon_{1.25} = \sqrt{\varepsilon_1 \varepsilon_2} = \sqrt{\frac{N_{sum}}{A \rho_1 \rho_2 \Delta t}}$$

This is equation (1).

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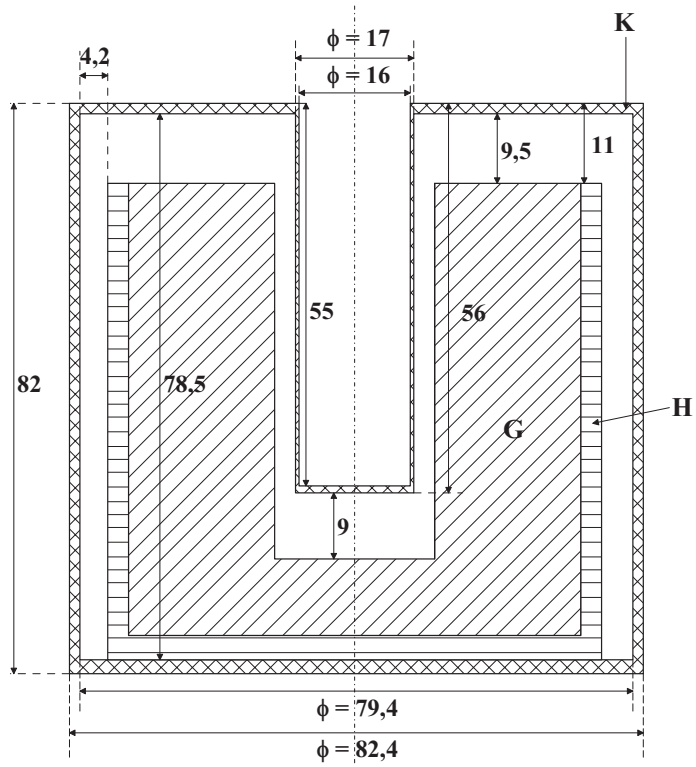


Figure 1

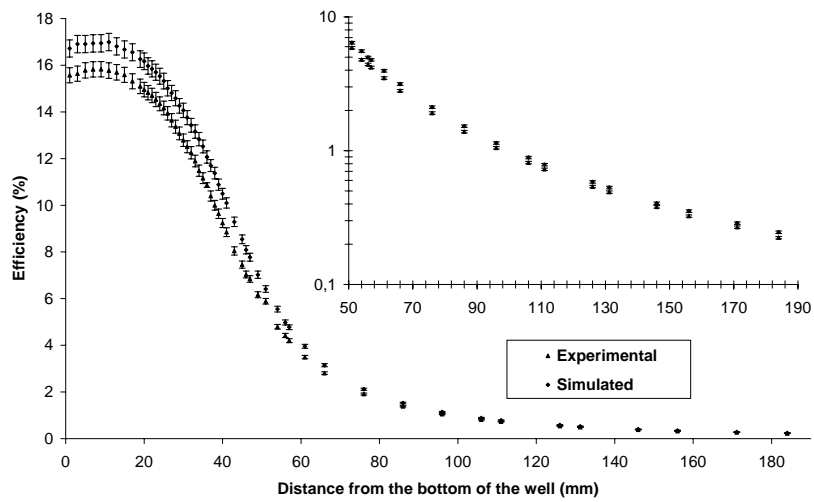


Figure 2

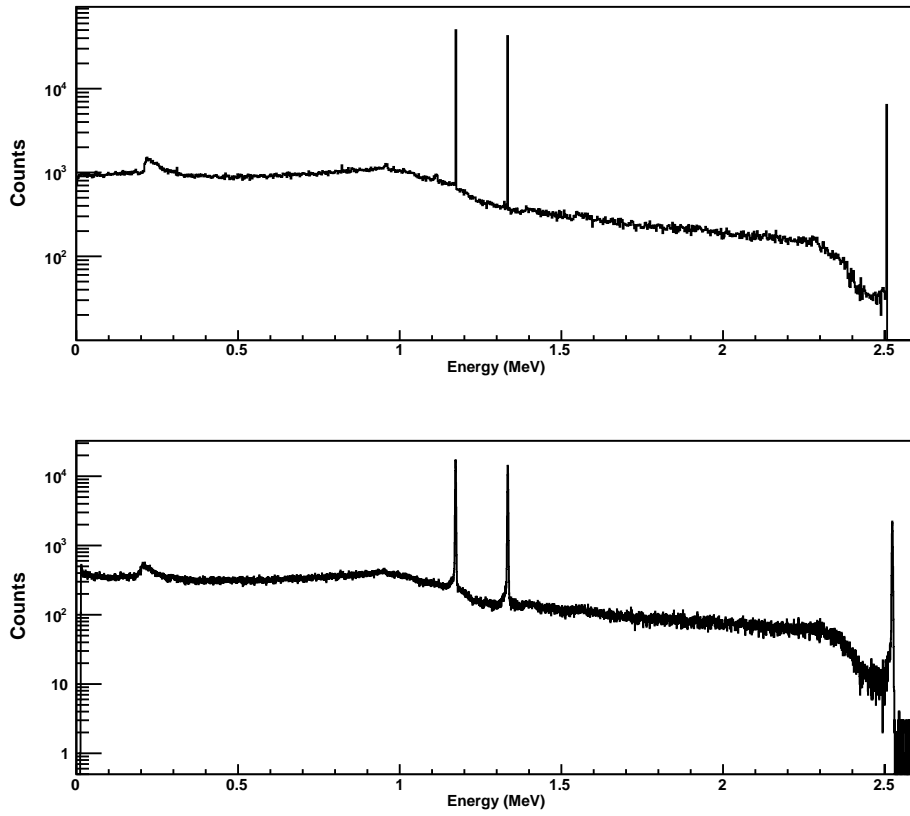


Figure 3

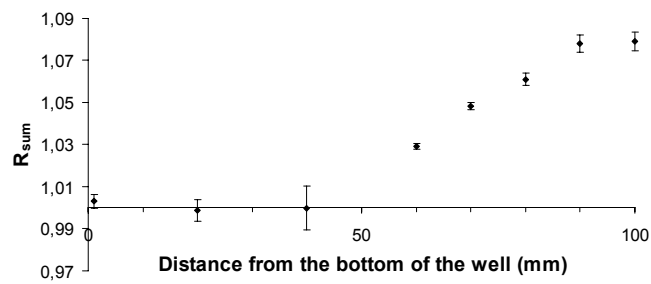


Figure 4